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BIODEGRADABLE FILM LAMINATE AND BIODEGRADABLE LABEL
[SEIBUNKAISEI FUJIRUMU SEKISOUTAI OYOBI SEIBUNKAISEI RABERU]

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SCOPE OF THE PATENT CLAIMS

CLAIM 1

A biodegradable film laminate characterized by the metal film layer (A) being a biodegradable film laminate formed through the anchor agent layer (B) on the biodegradable film layer (C), and the above anchor agent layer includes aliphatic polyester wherein the mol ratio (L/D) of an L - lactic acid residue and the L - lactic acid residue is 1-9, and the reduction viscosity is 0.3-1.0dl/g.

CLAIM 2

The biodegradable film laminate of claim 1 characterized by that the aliphatic polyester possesses a reactive group and polar group and the total density of these quantities are a 100-500 chemical equivalent /106g.

CLAIM 3

The biodegradable film laminate of claim 2 characterized by that the reactive group or the polar group is a hydroxyl group, epoxy group, amino group, carvone acid group, sulfone acid group, and phosphonic acid group and is among at least one kind of the of these salts.

CLAIM 4

The biodegradable film laminate of any of claims 1-3 characterized by that the aliphatic polyester has been

bridged with an isocyanate compound, epoxy resin, and at least more than one kind of formaldehyde resin.

CLAIM 5

The biodegradable film laminate of any of claims 1-4 characterized by that the biodegradable film layer consists of a poly lactic acid system film.

CLAIM 6

The biodegradable label where the biodegradability film laminate of any of claims 1-5 has been used as a support.

DETAILED DESCRIPTION OF THE INVENTION

[0001]

TECHNICAL BACKGROUND OF THE INVENTION

The present invention relates to a biodegradable film laminate, more specifically, it relates to the biodegradable poly lactic acid film laminate where a film layer possessing a metallic luster has been provided, even more so, it relates to the biodegradable label where a biodegradable film laminate possessing the above metallic luster has been used as a support, for example, for labels such as stickers or industrial brand names.

[0002]

CONVENTIONAL ART

Due to increased awareness in recent environmental problems, natural material or the development of product

where biodegradable configuration materials are used is performed flourishingly. Especially, the demand for molded articles that have used ingredient solution-related plastics are prolonged. In general plastic casts, there are many cases where various labels for the purpose of instruction and brand name descriptions, and for design-related improvements are adhered to the surface. However, in cases where stickers, brand names, and labels consisting of biodegradable plastic resin are attached there is the problem of only stickers, brand names and labels comprised of non-biodegradable plastic resin not decomposing and remaining in the soil.

[0003]

Its purpose is designated for design characteristics and visibility improvement on the likes of brand names and label film, and there are many cases where plastic films having a metal film layer are generally used as a support, and a polyethylene terephthalate film thickness of 200um degrees is usually used for plastic film at 25µm. However, in consideration of environmental problems such as the above, a biodegradable label has been suggested (Japanese Patent Laid-Open No. H7-44104 Gazette). According to this bulletin, polyester that is a biodegradable resin-based resin; natural macromolecules, microbe production plastic

and poly lactic acid system resin are used as substrate film. It is desirable to use poly lactic acid biological decomposition resin as a substrate film for labels due to its processing, productivity, various dynamic characteristics, and printing adaptability sides as a biodegradable resin.

[0004]

However, in cases where the metal film layer is molded as a poly lactic acid film, when general vacuum deposition occurs, bad coherency with the poly lactic acid film and the metal film layer is clearly indicated. Especially, when the biodegradable film having a metal film layer was used as a substrate for labels, after the adhesion processing, printing, protective film laminating, punching process after processing, and after actually being adhered as a label, a delaminating malfunction occurred between the biodegradable film and the metal film layer due to a deterioration over time.

[0005]

Conventionally, there is a method in which surface treatment for the film is done by the likes of a sputtering and corona process as a means to improve the coherency between the metal film layer and plastic film. Sputtering is disadvantageous on a productivity and cost side, and, in

the case of poly lactic acid film, the corona processing provides almost no coherency improvement effect at all.

[0006]

In addition, there is also a method which uses an anchor agent consisting of a poly urethane type resin and polyester-based resin, and a 2 liquid hardening type anchor agents as an external means to improve coherency between the metal film layer and plastic film, but when a poly lactic acid film is used a coherency improvement effect cannot be expected.

[0007]

PROBLEM TO BE SOLVED BY THE INVENTION

The main purposes of this invention are to raise adhesive strength between layers and improve coherency between the metal film layer and plastic film, and to offer a biodegradable label which uses a film laminate high of natural explication, and moreover the said biodegradable film laminate as a support.

[0008]

MEANS TO SOLVE THE PROBLEM

The present invention is a biodegradable film laminate characterized by the metal film layer (A) being a biodegradable film laminate formed through the anchor agent layer (B) on the biodegradable film layer (C), and the

above anchor agent layer includes the resin group polyester where the mol ratio (L/D) of an L - lactic acid residue and the L - lactic acid residue is 1-9, and the reduction viscosity is 0.3-1.0dl/g. Said resin group polyester preferably possesses a reactive group and a polar group, and as for the total density of these quantities a 100-500 chemical equivalent /106g is suitable.

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[0009]

Here, the aliphatic polyester reactive group or the polar group is preferred to be a hydroxyl group, epoxy group, amino group, imino group, a carvone acid group, a sulfone acid group, phosphonic acid group and at least one kind of these salts. And, an isocyanate compound, epoxy resin, and at least more than one kind of formaldehyde resin ideally bridge the aliphatic polyester.

[0010]

Moreover, a poly lactic acid film is suitable as a biodegradable film, the other forms of the present invention are the biodegradable labels, which had used the biodegradable film laminate as a support.

[0011]

DESCRIPTION OF THE PREFERRED EMBODIMENT

The present invention is a biodegradable film laminate where the metal film layer (A), for example, metal vapor deposition tapetum was formed through the anchor agent layer (B) on the biodegradable film layer (C).

[0012]

Here, the anchor agent layer (B) includes aliphatic polyester where the mol ratio (L/D) of L - lactic acid residue and D - lactic acid residue is 1-9, and the reduction viscosity is 0.3-1.0dl/g.

[0013]

Applying the anchor agent wherein aliphatic polyester was dissolved in a non-Halogen system organic solvent on the biodegradable film layer forms the anchor agent layer used for the present invention.

[0014]

The aliphatic polyester used for an anchor agent is based on lactic acid and is polymerized and provided. Lactic acid residue preferably contains 60 mol % or more, more preferably 70 mol % or more, furthermore preferably 80 mol % or more, particularly preferably 90 mol % or more, and most preferably 95 mol % or more. When a lactic acid residue is less than 60 mol %, good adhesion strength, and good biodegradability are not provided. In addition, it is preferred for the content of the aliphatic polyester lactic

acid residue to be less than 99.99 mol % from an aspect of the adhesive property.

[0015]

In addition, as for the mass, an aliphatic polyester with a lactic acid residue of more than 80 % mass is desirable, of more than 90 % mass is even more desirable, and of more than 95 % mass is especially even more desirable.

[0016]

The above aliphatic polyester can include lactic acid and other oxy acid co-polymers or gal Bonn acid and a polyester block consisting of a diol in a polymer molecule. As oxy acids for other than lactic acids which are copolymerizable, hydroxyacetic acid, 2-hydroxy isobutyric acid, 3-hydroxy butyric acid, 16 - hydroxy hexadeconic acid, 2-hydroxy - methyl butyric acid, 12 - hydroxy stearic acid, 4-hydroxy butyric acid, ten - hydroxy stearic acid, malic acid, citric acid, and gluconic acid are given. In addition, ester in the molecule of the hydroxy acid such as caprolactone, and water molecules from a - oxy acid such as lactide are lost and the created ringed ester is also used.

[0017]

Succinic acid, adipic acid, azelaic acid, and sebacic acid are nominated as gal Bonn acid, and ethyleneglycol, diethylene glycol, propylene glycol, and butanediol are

nominated for diol. In addition, the calculation (mol %) of the content of lactic acid when polyester consisting of diol and gal Bonn acid are co-polymerized is calculated as each of the individual units of gal Bonn acid, and diol. In addition, for the poly glycerin as a polyol ingredient, it is calculated with the number of the mols calculation included.

[0018]

The mol ratio (L/D) of L - lactic acid and D - lactic acid of the lactic acid residue in the aliphatic polyester is preferred to be in the range of 1-9. When L/D is smaller than 1, in other words, if D - lactic acid is in surplus then there is a rise in cost. In addition, when L/D exceeds 9, dissolubility for the general-purpose solvent of the non-Halogen system deteriorates, and the anchor agent gets coated and becomes difficult.

[0019]

The mol ratio of the L - lactic acid residue and D - lactic acid residue of polyester is requested from charged amount, but by using an optical rotation photometer (Horiba, Ltd. SEPA-200) confirmed that the mol ratio of the L - lactic acid residue and D - lactic acid residue of polyester was the same the one that was decided.

[0020]

Ether system solvents such as methyl ethyl ketone, methyl isobutyl ketone, and ketone system solvents such as the cyclohexanone, toluene, and aromatic series system solvents such as the xylene, ethyl acetate, butyric acid propyl, and ester system solvent tetrahydrofuran such as the butanoic acid, and diethyl ether are raised with the non-Halogen system solvent dissolving the anchor agent.

[0021]

As for the reduction viscosity of the aliphatic polyester, a 0.3-1.0dl/g is preferable. When reduction viscosity is less than it 0.3-0.1dl/g, cissing occurs at the time of coating, and a lack of adhesion strength occurs. In addition, viscosity of the coating liquid becomes higher when reduction viscosity exceeds 1.0dl/g, and coating fitness deteriorates.

[0022]

The reduction viscosity is a sample density of 0.125g/25ml and the measurement solvent is a chloroform measurement temperature of 25 degrees Celsius, are the values measured by using a ubbelohde viscosimeter

[0023]

As for the glass transposition temperature (hereinafter referred to as T_g) of the aliphatic polyester, a range of 35-60 degrees Celsius is preferable. When the adhesion

strength of the metal film layer becomes insufficient, and T_g exceeds 60 degrees Celsius when T_g is less than 35 degrees Celsius, cracking occurs easily in the metal film layer and is inferior to design characteristics and gas barrier characteristics. The favorable lower limit of T_g is 38 degrees Celsius, and a more favorable lower limit is 40 degrees Celsius. In addition, the upper limit of T_g is 57 degrees Celsius, and here, where a more favorable upper limit is 55 degrees Celsius, a 10mg sample is taken from an aluminum pan, covered with an aluminum lid, strongly crimped, and is the value measured by the speed of the rise in temperature of 10 degrees Celsius/by the DSC (Differential Scanning Calorimeter) method.

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[0024]

An aliphatic polyester possessing a reactive group or a polar group is preferable, and a total density in a range of 100-500 chemical equivalent /106g for these is suitable in this case.

[0025]

If the total density of the reactive group and polar group is less than a 100 chemical equivalent /106g, for example, coherency with the metal evaporation coating is not provided, then heat resistance deteriorates without

sufficiently reacting with cross-linker (bridging agent) at the time of bridging, and heat resistance of around 50-60 degrees Celsius becomes hard to use for the required use. In addition, water resistance deteriorates when a 500 chemical equivalent /106g is exceeded.

[0026]

Here, the reactive group reacts with another functional group and is a functional group that can form covalent bonds; for example, a hydroxyl group, the epoxy basis, amino group, imino group, and a carvone acid group are given. In addition, for example, a hydroxyl group, amino group, imino group, carvone acid group, sulfone acid group, phosphonate acid group, and these salts are nominated for the polar group. As the salt amino group, it is Halogen salt with chlorine, bromine in the case of the imino group, or acetate, and in the case of carvone acid group, sulfone acid group, potassium, alkali metal salts such as the sodium and an ammonium salt are given.

[0027]

In these reactive groups or polar groups, a hydroxyl group is preferable at the point where high coherency is provided. The density of these functional groups can be measured by a calculation from the charged amount or well-known methods such as the titration.

[0028]

In addition, using a hydroxyl group as an example, When describing the request of the density of the reactive group, the density of the hydroxyl group measures acid value in a value (calculated from polyol and a hydroxyl group value of the poly glycerin added to the reaction) calculated from the charged amount and the provided acid value (acid value by the resolution of the lactide = Hydroxyl group value by the resolution of the lactide) may add and request, moreover, a resin hydroxyl group where surplus phenyl isocyanate is added and reacted and next, non-reaction phenyl isocyanate is reacted with surplus diethyl amine, and the amount of non-reaction diethyl amine can be found by titrimetry where titration is performed by acid.

[0029]

The methods to coordinate these reactive groups in the appointed density range, for example, in the case of a hydroxyl group, are in the following methods. For example, a method in coordination with molecular weight of the polyester, a polymerization method for poly lactic acid with lactide, or a method to add a polyol compound in polymerizing, a method to add polyol after poly lactic acid polymerizing, and to do de-polymerization, hydroxy basis component epoxy compounds and a method to react it, and a

method to introduce plural hydroxyl groups into the distal end are given.

[0030]

Poly glycerin, polyvinyl alcohol, sorbitol, glucose, galactose saccharide, pentaerythritol are nominated for polyol. Poly glycerin is preferable in/among these.

[0031]

As for the polymerizing degree of the poly glycerin, 3-20 is preferable and more than 5 is more preferable, and preferably the upper limit is equal to or less than 15. At/With a polymerizing degree of less than 3, coherency to the good metal film layer is not provided. In addition, when polymerizing degrees of the poly glycerin exceed 20, water resistance turns worse.

[0032]

As for the content of the poly glycerin, less than 20% mass among the aliphatic polyester is preferable, and more preferable is less than 10% mass, and even more preferable is less than 10% mass, and particularly preferable is less than 3% mass. In addition, the content of the poly glycerin is 0.01% mass or more, preferably 0.05% mass or more, furthermore preferably is 0.1% mass or more, and particularly preferable is a 0.02% mass or more in the aliphatic polyester.

[0033]

When content of the poly glycerin is beyond 20% mass, water resistance turns worse. And, quantity of adhesion of the metal film layer falls with less than 0.01% mass.

[0034]

With methods to introduce reactive groups other than the hydroxyl group or the polar group, a method to do ring-opening polymerization of lactide in the presence of poly arylamine, poly methacryl amine, poly - N - ethyl Amino acrylate, and diethnol amino are nominated for an example in case of the amino acids. In addition, for an example in case of the imino group, there is a method that does ring-opening polymerization of lactide in the presence of polyethylene imine. In addition, for an example of a carvone acid group case, there is a method to do ring-opening polymerization in the presence of poly (meta) acrylic acid. Furthermore, a method to do ring-opening polymerization of lactide in the presence of sulfo-isophthalic acid is nominated for an example in case of the sulfone acid group.

[0035]

In the present invention, the manufacturing method of aliphatic polyester is not limited in particular and conventional well-known methods can be used. For example,

methods for heat ring-opening polymerization where the lactide which is the dimer of the lactic acid and other oxy acid are dissolved and mixed, and uses a well-known ring-opening polymerization catalyst (for example, octyl acid tin, aluminum acetylacetonate), and a method to perform direct dehydration poly-condensation by heating and decompression are given

[0036]

In the present invention, multifunctional isocyanate, multifunctional epoxy, melamine resin, penzolkuanamine resin, cross-linkers such as the formaldehyde resin, a viscosity regulator, a deterioration inhibitor, and the likes of coloring agents can be combined a with an anchor agent if necessary,

[0037]

The isocyanate of a aliphatic system is preferable especially as a cross-linker. As for aliphatic isocyanate here, hexamethylene-diisocyanate, tolidine diisocyanate, hydrogenation toluylenediisocyanate, hydrogenation diphenylmethane diisocyanate and the likes are given, and 3 quantities of these are particularly preferable.

[0038]

In addition, the anchor agent, from a point of view promoting biodegradability, contains more than 70% mass in

the aliphatic polyester in the solidity, furthermore, more than 80% mass, and especially more than 90% mass is preferable.

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[0039]

The an anchor agent is applied to a biodegradable film layer, preferably to a poly lactic acid film, forming the anchor agent layer. The application method of the anchor agent is not limited in particular, and well-known methods such as reverse roll coater, gravure coater, micro-gravure coater, comma coater, bar coater, air doctor coater are used.

[0040]

The film thickness of the anchor agent layer is not limited in particular, but less than 2.0 μm are preferable at more than 0.01 μm . When the film thickness of the anchor agent layer is less than 0.01 μm , the film and the coherence strength of the metal film layer are not sufficiently provided, and a minute crack occurs in the metal film layer when metal film layer is formed by vacuum deposition if it is over 2.0 μm , and there are case where it becomes unfavorable in appearance. In addition, there is a gas barrier characteristic deteriorate when it is used as a casing.

[0041]

In addition, the anchor agent is coated before the extension of the biodegradable film, and even an in-line coating extension or an offline coating after extension by another process is good afterwards to coat the anchor agent on the biodegradable film.

[0042]

As for the biodegradable film layer (C), suitably a poly lactic acid film is used, but a poly - L - lactic acid film, especially, one where the optical purity of the L - lactic acid residue content is more than 97 mol % is preferable, and even more preferable is one that is more than 99 mol %.

[0043]

As for the biodegradable film laminate of the present invention, the anchor agent layer (B) is laminated by the biodegradable film layer (C), and the metal film layer is formed on the top. And preferably the metal film layer is formed with the metal vapor deposition layer having a metallic luster. And for the metal vapor deposition layer having a metallic luster, aluminum, gold, silver, copper, chrome, nickel, tin, titanium and these alloys are given, but it is not particularly limited to these. Aluminum is nominated as a cheap metal vapor deposition layer. The vacuum deposition method and the sputtering method are

nominated as the metal vapor deposition method, and it is not limited if it is this method in which the metal in the molecular shape and a cluster is laminate on the biodegradable film layer (C).

[0044]

10nm~100nm is preferable for the thickness of the metal film layer (A) and with less than 10nm, enough metallic luster cannot be acquired, and there are times when it is undesirable as a label material. In addition, as a casing there are times when the gas barrier characteristic deteriorate and it is undesirable. However, it is evaporated to less than 10nm, and there are also times when the design nature is shown as a half mirror form, and the thickness of the metal film layer is not limited in particular. In addition, not only do the film and the coherence strength of the metal film layer deteriorate when the metal film layer exceeds 100nm, but there is also no merit for costs.

[0045]

The biodegradable film can produce a biodegradable label by using a laminate. After natural rubber and terpene-based resin or adhesiveness such as poly lactic acid (system) resin and biodegradable rubber or an adhesive layer with resin is formed on the side of the metal film

layer of the biodegradable film laminate or the other side of the metal film layer, the detached sheet is laminated. You may laminate to a biodegradable film laminate comprising of the metallic luster after having applied an adhesive layer on the detached sheet beforehand. Since the detached sheet is disposed after the use of the label, preferably, after the disposal of biodegradable plastic or paper the use of the material, which does not give the natural environment a load, is expected, but even ones that are not particularly biodegradable are separable when the label is used.

[0046]

Moreover, the biodegradable film laminate of the present invention, especially poliomyelitis lactic acid film laminate, sets a heat seal layer, and applies the use as a casing of bag-shaped food and the likes or an adhesive or glue, and can be used as a seal, a label, a sticker and a tag.

[0047]

In addition, the print layer and the outside coat layer can be interposed between the biodegradable film layer (C) and the anchor agent layer (B) and on the metal film layer (A) of the biodegradable film layer (C) and the other side. In addition, by carrying out the coloration in the anchor

agent layer and the other coat layer, the design nature can be increased by adjusting color tones such as gold evaporation and copper evaporation and colorizing even if the metal film layer of aluminum evaporation is formed, if it does not physically deteriorate even if the print ink and coat layer are left in the natural environment, not being limited in particular. However, when a film of the poly lactic acid system is used for the biodegradable film layer, it is preferable to use a poly lactic acid ink and coating from a coherency and biodegradability aspect. In addition, the easily adhered coat may be arranged on the surface of the biodegradable film layer to improve coherency with the biodegradable film layer and the print layer. Furthermore, laminating of film on the surface of the print layer can be done to improve the weathering ability of the print layer and abrasion characteristics-resistant.

[0048]

EMBODIMENT

The present invention will be explained in detail based on an following embodiments, but it is not limited to these.

(1) Production of the aliphatic polyester

Embodiment A

1,000 parts of DL lactide, 10 parts of Poly glycerin (Daisel chemistry PGL10: Hydroxyl group density 850KOHmg/g) which is polymerization degree of 10, and aluminum acetylacetonate 1 part as a ring-opening polymerization catalyst, were charged in 4 low flasks, under a nitrogen atmosphere, ring-opening polymerization is done by heat-melting at 180 degrees Celsius for three hours, and polyester (I) is acquired by removing the residual lactide under vacuum by evaporation.

[0049]

To find the hydroxyl group density of polyester (I) logically, the hydroxyl group density of polyester (I) is selected only in the hydroxyl group of the poly glycerin origin. In addition, the acid value also becomes 0. However, actually, in the raw materials the inclusion of impurities is common, and in this case, lactyl lactic acid included in DL lactide holds the most of impurities. In

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this case the lactyl lactic acid of impurities acts as a polymerism start agent in substitution for poly glycerin, there are times when a hydroxyl group that should not occur without impurities does occur. In addition, in the lactyl lactic acid which is oxy acid, since the acid value and the hydroxyl group value are equivalent, the hydroxyl group

density of impurities origin can know it by measuring the acid value of polyester (I). When the hydroxyl group density of poly glycerin and the measured acid value is added, the requested aliphatic polyester hydroxyl group density is acquired.

[0050]

Based on the method mentioned above, the hydroxyl group density of polyester (I) was done as follows and requested. The hydroxyl group density of glycerin origin of polyester (I) is a calculation $(850 \times 1,000) / 56 \times 10 / (1,000 + 10) = 150$ based on (KOH conversion hydroxyl group density / molecular weight of KOH X mass portion of the poly glycerin / mass portion of the polymer), and was a 150 chemical equivalent /106g. In addition, the hydroxyl group density of impure origin, namely, the measurements of the acid value of the polyester (I) was over a 40 chemical equivalent /106g. Therefore, the hydroxyl group density of polyester (I) was found to be a 190 chemical equivalent /106g of the value in total. In addition, 0.2g of acid value polyester (I) was dissolved in 25ml of chloroform and measured with KOH a 0.1N ethanol solution. A phenolphthalein was used as an indicator of the titration.

[0051]

Embodiment B

1,000 parts of DL lactide, 5,6 parts of poly glycerin (Daisel chemistry PGL10: Hydroxyl group density 850KOHmg/g) which is a polymerization degree of 10, and aluminum acetylacetonate 1 part as a ring-opening polymerization catalyst, were charged in 4 low flasks, under a nitrogen atmosphere, ring-opening polymerization is done by heat-melting at 180 degrees Celsius for three hours, and polyester (II) is acquired by removing the residual lactide under vacuum by evaporation.

[0052]

Here, when the hydroxyl group density of polyester (II) like embodiment 1 was found, the hydroxyl group density of poly glycerin origin were 85 and a chemical equivalent /106g, and hydroxyl group density of impurities origin 25 and a chemical equivalent /106g. Therefore, the hydroxyl group density of polyester (II) is a 110 value chemical equivalent /106g in total.

[0053]

Embodiment C

1,000 parts of DL lactide, 16.5 parts of poly glycerin (Daisel chemistry PGL10: Hydroxyl group density 850KOHmg/g) which is a polymerization degree of 10, and aluminum acetylacetonate 1 part as a ring-opening polymerization catalyst, were charged in 4 low flasks, under a nitrogen

atmosphere, ring-opening polymerization is done by heat-melting at 180 degrees Celsius for three hours, and polyester (III) is acquired by removing the residual lactide under vacuum by evaporation.

[0054]

Here, when the hydroxyl group density of polyester (III) like embodiment 1 was found, the hydroxyl group density of poly glycerin origin were 246 and a chemical equivalent /106g, and hydroxyl group density of impurities origin 30 and a chemical equivalent /106g. Therefore, the hydroxyl group density of polyester (III) is a 110 value chemical equivalent /106g in total. Therefore, the hydroxyl group density of polyester (III) is 276 value chemical equivalent /106g in total.

[0055]

Embodiment D

1,000 parts of DL lactide, 16.5 parts of poly glycerin (Daisel chemistry PGL10: Hydroxyl group density 850KOHmg/g) which is a polymerization degree of 10, and aluminum acetylacetonate 1 part as a ring-opening polymerization catalyst, were charged in 4 low flasks, under a nitrogen atmosphere, ring-opening polymerization is done by heat-melting at 180 degrees Celsius for three hours, and

polyester (IV) is aquired by removing the residual lactide under vacuum by evaporation.

[0056]

Here, when the hydroxyl group density of polyester (IV) like embodiment 1 was found, the hydroxyl group density of poly glycerin origin were 409 and a chemical equivalent /106g, and hydroxyl group density of impurities origin 50 and a chemical equivalent /106g. Therefore, the hydroxyl group density of polyester (IV) is a 110 value chemical equivalent /106g in total. Therefore, the hydroxyl group density of polyester (IV) is 459 value chemical equivalent /106g in total.

[0057]

Composition of polyester (I) - (IV) provided in embodiment A~D and properties of matter are shown in table 1. The specifications shown in table 1 and the measurement of properties of matter were carried out according to the next method.

[0058]

<Reduction viscosity> sample density 0.125g /25ml, measurement solvent chloroform, and measurement temperature 25 degrees Celsius, were measured with an Ubbelohde viscosimeter.

[0059]

<Glass transition point (T_g) > glass point move point
(T_g) was measured by DSC method.

[0060]

The lactic acid residue during lactic acid system
polyester composition<lactic acid residue (mass %)>
prescribed the charged amount and it is shown in table 1.
However, it was confirmed that the same quantity was
included in polyester by NMR of 500MHz.

[0061]

The mol ratio of the L - lactic acid residue and D -
lactic acid residue of the mol ratio> polyester of the <L -
lactic acid residue and the D - lactic acid residue, were
requested from the charged amount, but it confirmed that
they were the same mol ratio of the L - lactic acid residue
and D - lactic acid residue in polyester that was decided
by/ with rotation photometer (Horiba, Ltd. SEPA -200).

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[0062]

Table 1

Polyester composition of the anchor agent

	Embodiment A	Embodiment B	Embodiment C	Embodiment D
Polyester	I	II	III	IV

Lactic acid residue (% by weight)	99.0	99.4	98.4	97.4
LD (mol ratio)	1	1	1	1
Reduction viscosity (dl/g)	0.49	0.74	0.50	0.33
Hydroxyl group density (chemical equivalent /106g)	190	110	276	459
T _g ()	49	49	49	48
Poly glycerin segment polymerization degree	10	10	10	10

[0063]

(2) The production of the biodegradable film laminate

Embodiment 1

1 poly lactic acid resin (polyester (I)) mass portion and coater consisting of 40 toluene mass portions was applied to one side of the poly lactic acid film of thickness of 25um as a biodegradable film by/with a gravure coater so that the dry film thickness became 0.02um and formed the anchor agent layer. Then, a metal film layer of a 45nm thickness was formed by aluminum vacuum deposition.

[0064]

Embodiment 2

1 poly lactic acid resin (polyester (II)) mass portion and coater consisting of 36 toluene mass portions and 4 cyclohexanone mass portions was applied to one side of the poly lactic acid film of thickness of 25um as a biodegradable film by/with a gravure coater so that the dry film thickness became 0.02um and formed the anchor agent layer. Then, a metal film layer of a 45nm thickness was formed by aluminum vacuum deposition.

[0065]

Embodiment 3

As biodegradable film, a coater consisting of 1 poly lactic acid resin (polyester (I)) mass portion, 0.1 aliphatic isocyanate (Brand name: Duranet TPA -100, product made by Asahi Chemical Industry Co.,Ltd. Corporation) mass portion, 0.02 dibutyl tynedilaurate [TN: *Possibly Miss-*

spelled] (a product made in Tokyo Kasei Kogyo Corporation Co., Ltd.) mass portion, and 40 toluene mass portion were applied to/on 1 side of the poly lactic acid film of a thickness of 25um by gravure so that the dry film thickness would become 0.02um and formed the anchor agent layer.

[0066]

Embodiment 4

Except when it was not the poly lactic acid resin polyester (III), a laminate comprising aluminum (thin) film was obtained the same as it was in Embodiment 3

[0067]

Embodiment 5

Except when it was not the poly lactic acid resin ester (IV), a laminate comprising aluminum (thin) film was obtained the same as it was in Embodiment 3

[0068]

Comparison example 1

A coating liquid (coater) consisting of 1 mass portion of polyester (Brand name: Byron 200, product made by TOYOBO Co., Ltd. Corporation), 0.1 mass portion of an isocyanate compound (Brand name: Coronet L, product made by Japan Polyurethane Co., Ltd. Corporation), 18 toluene mass portions, 18 methyl ethyl ketone mass portions, and 4 cyclohexanone mass portions was applied to one side of the

poly lactic acid film of thickness of 25um as a biodegradable film by a gravure coater so that the dry film thickness became 0.02um and formed the anchor agent layer. Then, a metal film layer of a 45nm thickness was formed by aluminum vacuum deposition.

[0069]

Comparison example 2

Corona processing was performed on one side of the poly lactic acid film of thickness 25um as a biodegradable film, then, a metal film layer of a 45nm thickness was formed by aluminum vacuum deposition.

[0070]

Comparison example 3

A 45nm direct film thickness metal film layer was formed by aluminum vacuum deposition as a biodegradable film on one side of the poly lactic acid film of having a thickness of 25um.

[0071]

The performance evaluation of the biodegradable film laminates provided in the embodiment and the comparison examples were performed by the next method.

[0072]

<Coherence strength>

For the measurement of the coherence strength of the biodegradable film layer and the metal film layer, at first, an adhesive (an adhesive in which 100 mass portions of AD - 122 made in Orient Morton Co., Ltd. Corporation that is mixed with 5 mass portions of CAT -10) was applied to a 25 μ m thick polyester film so that the dry film thickness would become 3 μ m. Dry laminating was done so that the sample aluminum evaporated surface of each biodegradable film laminate became the adhesion side on the surface, making a specimen, then after having aged at 40 degrees Celsius this specimen was measured by the 180 degrees detachment method for 24 hours. The unit of measurements is g/15mm.

[0073]

<Detachment strength: Water resistance>

The sample of the predetermined dimensions from the biodegradable film laminate provided in the embodiment and comparison examples were compiled, this sample was dipped in water of 25 degrees Celsius, and it was taken out 1 hour later and the aluminum evaporated surface was rubbed with a finger.

The evaluation standard is as follows.

: Evaporation layer does not come off.

○: Part of the evaporation layer came off.

: Most of evaporation layer came off.

X: The evaporation layer came off only by a dip.

<Detachment strength: Heat-resistant> an aluminum evaporated surface of the biodegradable film laminate provided in the embodiment and the comparison examples for a hot plate of 80 degrees Celsius posted was positioned so it would be on top and rubbed against the evaporated surface with a cloth.

The evaluation standard is as follows.

:The evaporation layer does not come off.

○:A part of the evaporation layer came off.

: The half-degree of the evaporation layer came off.

X: The most of evaporation layer came off.

[0074]

Table 2

		Embodiment	Embodiment	Embodiment	Embodiment	Embodiment	Comparison	Comparison	Comparison
		1	2	3	4	5	Example 1	Example 2	Example 3

Bio Fil m lam ina te	Biodegra dable film Material s thicknes s (um)	poly lacti c acid 25	poly lacti c acid 25	poly lacti c acid 25	poly lacti c acid 25	poly lacti c acid 25	poly lacti c acid 25	poly lacti c acid 25	poly lactic acid 25
	The anchor agent layer Material s thicknes s (um)	Polye ster (I) 0.02	Polye ster (II) 0.02	Polye ster (I) 0.02	Polye ster (III) 0.02	Polye ster (IV) 0.02	Polye ster Resin A 0.02	--	--

	The metal film layer Material s thickness (um)	45	45	45	45	45	45	45	45
Properties of material	Coherence strength (g /15mm)	450	400	450	440	460	320	15	30
tereva luation	debond interfac e	AI//P ET	AI//P ET	AI//P ET	AI//P ET	AI//P ET	PLC// AI	PLC// AI	PLC//AI
tio n	Biodegradability	○	○	○	○	○	X	○	○
	Water resistance				○				

heat-resistance									
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[0075]

The above evaluation result is shown in table 2.

In the column of the detachment interface of table 2, A1 is a aluminum film layer, The PET is the anchor agent layer (polyester), and PLC means the biodegradable film layer (the poly lactic acid film layer).

[0076]

In regards to the evaluation result of the detachment strength, whereas the specimen of the comparison example from table 2 exfoliated with the aluminum evaporated layer and ingredient solution-related film layer (the poly lactic acid film layer) entirely, all the specimens of the embodiment exfoliated with the aluminum evaporated layer and the polyester film layer.

[0077]

The needs assessment of the biodegradability went by the next method. A specimen of predetermined dimensions from a biodegradable film laminate provided in the embodiment and comparison examples were made. An adhesive layer was formed on the aluminum evaporation surface of the specimen by a comma coater so that the natural rubber and terpene-

based resinous mixture would become a 30g/m² mass after drying. Mass after the drying is 30g/m² I form an adhesive layer. In one way, the adhesive layer and the silicone layer with the detachment paper that silicone layer arranged on 1 side are laminated to put them together. Furthermore, the print layer on the surface of the poly lactic acid film (biodegradable film) is formed and makes the biodegradable label. In a process to make a biodegradable label it is not necessary to exfoliate in the interface of the poly lactic acid film layer and the aluminum evaporated layer, moreover, the biodegradable label is adhere to the biodegradable resin ejection cast after having removed the detachment paper and buried it in the soil at a depth of 10cm.

[0078]

Except for comparison example 1, the embodiment and the other comparison examples identified the decomposition as the label and the ejection cast six months later.

[0079]

It should be understood that all the forms of the enforcements disclosed this time and the embodiments are illustrations, and they are not restrictions . The range of the present invention is shown according to the SCOPE OF THE PATENT CLAIMS and not the above description; it is

intended to include an equal meaning to the SCOPE OF THE
PATENT CLAIMS and all changes within the range of this
patent.

[0080]

EFFECT OF THE INVENTION

As explained above, the biodegradable polyester,
especially, the biodegradable laminating film laminate
which laminated the metal film layer on a biodegradable
laminating film through the anchor agent layer of the poly
lactic acid resin shows superior coherence strength between
the layers of the metal film layer and biodegradable film
layer. In addition, especially, the biodegradable label
which was made by using said biodegradable laminating film
stacks is superior in design characteristics, decomposition
characteristics, and does not create a burden for the
natural environment, and especially when it is used with an
ejection cast consisting of biodegradable plastic, it is
very useful.

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